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## Interdecadal changes of summer aerosol pollution in the Yangtze River Basin of China, the relative influence of meteorological conditions and the relation to climate change



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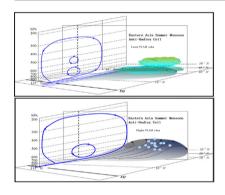
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#### HIGHLIGHTS

## The aerosol pollution was heavier in the M-LYR during the weak monsoon years and opposite in the strong monsoon years.

- The interdecadal changes in meteorological conditions and their associated aerosol pollution have experienced four periods since the 1960s.
- Among later three pollution increased periods, about 51%, 25% and 60% of the aerosol pollution change comes from the contribution of worsening weather conditions, which are found to be greatly affected by changes of EASM. Worsening weather conditions contributed 51%, 25%, and 60% respectively in the later three periods.

#### GRAPHICAL ABSTRACT



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## ABSTRACT

Winter is a season of much concern for aerosol pollution in China, but less concern for pollution in the summertime. There are even less concern and larger uncertainty about interdecadal changes in summer aerosol pollution, relative influence of meteorological conditions, and their links to climate change. Here we try to reveal the relation among interdecadal changes in summer's most important circulation system affecting China (East Asian Summer Monsoon-EASM), an index of meteorological conditions (called PLAM, Parameter Linking Air Quality and Meteorological Elements, which is almost linearly related with aerosol pollution), and aerosol optical depth (AOD) in the middle and lower reaches of the Yangtze River (M-LYR) in central eastern China during summertime since the 1960's. During the weak monsoon years, the aerosol pollution load was heavier in the M-LYR and opposite in the strong monsoon years mainly influenced by EASM and associated maintenance position of the anti-Hadley cell around 115°E. The interdecadal changes in meteorological conditions and their associated aerosol pollution in the context of such climate change have experienced four periods since the 1960's, which were a relatively large decreased period from 1961 to 1980, a large rise between 1980 and 1999, a period of

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slow rise or maintenance from 1999 to 2006, and a relatively rapid rise between 2006 and 2014. Among later three pollution increased periods, about 51%, 25% and 60% of the aerosol pollution change respectively come from the contribution of worsening weather conditions, which are found to be greatly affected by changes in FASM.

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#### 1. Introduction

The wintertime aerosol pollution in the central east of China has attracted much attention after the occurrence of persistent and severe aerosol pollution in January 2013 (Guo et al., 2014; Sun et al., 2014; Wang et al., 2014; Zhang et al., 2013). However, what was the aerosol pollution in summer and how did it change for a long time, and the relationship between this change and the interdecadal climate change in eastern Asia are only a few studies involved (Zhang et al., 2010). The most direct factor affecting the change of the air pollution is the weather conditions that closely related to the pollution (Leibensperger et al., 2008; Li et al., 2005; Wei et al., 2011; Zhang et al., 2009a, 2015; Zhong et al., 2017, 2018). Pollution-related meteorological conditions will also be affected by climate change. However, how the climate change, with an interdecadal shift of averaged climate condition as characters, affects the local and regional pollution-related metrological condition, it is still a question with large uncertain. How much of the long-term change in aerosol pollution in the summer is caused by climate-weather change, and how much of the influences from weather condition change is another particular concern issue.

East Asian Summer Monsoon (EASM) is considered to be the most important component of the climate system affecting the summer meteorological conditions in central eastern China. Many studies have shown that the EASM has been weakening since the end of the 1970s resulting in a "southern China flood and northern China drought" rainfall pattern (Hu, 1997a; Wang, 2001; Yu et al., 2004; Zhou et al., 2009b). To explore the long-term changes in pollution-related meteorological condition and the associated aerosol pollution in the summertime, and their relation to the EASM change, here we focused our study on summer aerosol pollution in middle and lower reaches of the Yangtze River (M-LYR) of China (110-125°E, 25-35°N), by analyzing the changes in aerosol optical depth (AOD), an index of meteorological conditions (called PLAM), and their links with long-term variation of EASM since 1961. The main purpose is to understand the interdecadal changes in summer aerosol pollution and relative influence from meteorological conditions in the M-LYR and their relation to east Asian climate change.

## 2. Data and method

## 2.1. Parameter Linking Air Quality and Meteorological Elements (PLAM)

In this paper, a PLAM index is used to describe the weather conditions that are most closely associated with aerosol pollution in the M-LYR. PLAM is an index extracted from the diagnosis and analysis of the connection between long-term (2000–2007) meteorological elements and PM2.5, which can linearly reflect the contribution of meteorological conditions to the change of PM mass concentration (Wang et al., 2012; Wang et al., 2013; Zhang et al., 2009b). A high value of the index represents meteorological conditions that are beneficial for the accumulative increase of pollution aerosols and transformation of secondary aerosols, called as "adverse meteorological conditions of pollution", conversely a low value indicates meteorological conditions conducive to the dilution of the pollution aerosol concentration. The first use of PLAM is to evaluate a relative contribution of weather conditions and reduction on air quality change during the Beijing Olympic in 2008 (Zhang et al., 2009b). Thereafter, PLAM was widely used to identify the contribution of specific meteorological factors to a 10 d haze-fog event in 2013 (Zhang et al., 2013), evaluate the relative influence of meteorological factors on changes in aerosol mass concentrations and chemical compositions in different regions of China during winter from 2006 to 2013 (Zhang et al., 2015) and estimate the feedback effect of weather conditions on the explosive increase in PM<sub>2.5</sub> mass concentration during accumulation stages in Beijing (Zhang et al., 2017; Zhong et al., 2017).

## 2.2. Aerosol optical depth (AOD)

550 nm AOD data from 1961 to 2005 is calculated based on observations of visibility and the water pressure from 504 key climate stations in Eastern China (Chen et al., 2008). These AODs information has been compared to the inversion AOD by MODIS satellite data (Elterman, 1970; Qiu and Lin, 2001), exhibiting good consistency and rationality (Li et al., 2011). The AOD data from 2006 to 2012 is from the aerosol remote sensing of network of China Meteorological Administration (CASNET) (Che et al., 2009).

# 2.3. East Asian Summer Monsoon (EASM) index derived on the basis of monsoon rainfall

By using NCEP/NCAR  $2.5^{\circ} \times 2.5^{\circ}$  reanalysis data (from 1961to 2014), the National Climate Center/China Meteorological Administration (NCC/CMA) Mei-Yu data, the vertical structures for the East Asian monsoon circulation and the characteristics of annual floods or drought were obtained in the M-LYR. For an accurate assessment of annual characteristics of drought in this area, the China Meteorological atmospheric precipitation observation reports for 1961–2010 were also employed (dot in Fig. 1b), as well as the hydrological observing stations in this area, including the water level and runoff observation data of Poyang Lake (Hukou Station) (29.4°N, 116.1°E) and Datong Station (30.7°N, 117.6°E) (Fig. 1b), prior to analyzing of these meteorological and hydrological data, the quality control and test was made (Niu et al., 2013).

Symbols and size of summer monsoon index could reflect the intensity of summer monsoon. By using 160 surface observation data of national meteorological center, a study on the relationship between widespread drought/flooding in the M-LYR and high-pressure in South Asia has been made (Zhang and Wu, 2001). The main structure of the summer precipitation in China in the past 50 years exhibited the inter-decadal precipitation changes in summer. The two important time node appear in 1978 and 1992, when years the dominant contribution was from equatorial Pacific tropical circulation due to the changes of the EASM system (Ding et al., 2008). Therefore, the index analysis based on of the monsoon on droughts and floods in the Yangtze River basin is the key to the summer winds. Analysis of drought/flood conditions in the Yangtze River region is the key to studying the impact of the summer monsoon. Furthermore, How to accurately analyze assess changes in annual monsoon rains in Yangtze River basin and flood or drought, to consider other complex factors in the watershed is necessary (Huang et al., 2008), such as geological features, the effect of lakes etc. study on annual summer monsoon intensity discrimination and recognition is very important (Li et al., 2011; Lin and Wang, 2002; Matsumoto, 1997). To determine the impact of monsoon on the atmospheric aerosol pollution is more important, because of aerosol scavenging effects significantly by monsoon rainfall (Che et al., 2007; Deng et al., 2014).

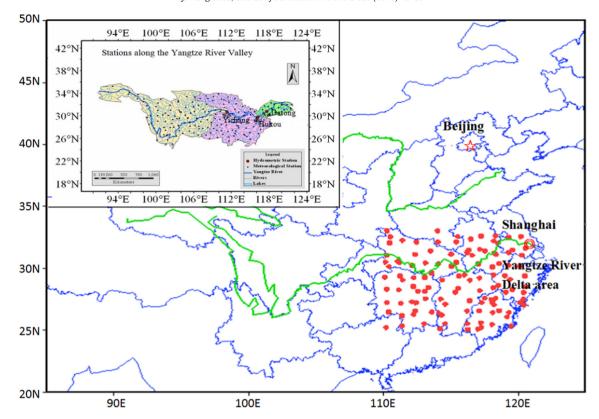


Fig. 1. Domain for PLAM (Parameter Linking Air Quality and Meteorological Elements) and the stations with AOD (aerosol optical depth) data estimated (red dots). The precipitation stations are also marked.

Based on the monsoon index reflecting the summer monsoon rainfall in the Yangtze River basin (Eq. (1), (Niu et al., 2013), this paper analyzing the evolvement of summer monsoon index series:

$$I_{(\sigma_{\gamma},\sigma_{\omega})} = \frac{1}{2} (\beta_{\lambda} + \beta_{w}) e^{-\frac{1}{2}(\sigma_{\gamma} + \sigma_{\omega})} + \sqrt{\beta_{\gamma}^{2} + \beta_{\omega}^{2}}$$
 (1)

where  $\beta_r$  and  $\beta_w$  are the inter-annual differences of observed atmospheric rainfall into the Yangtze River valley and watershed runoff respectively, calculated on the basis of historical data;  $\sigma_r$  and  $\sigma_w$  respectively represent the standardized atmospheric precipitation index within the watershed and the standardized runoff volume index of the key representative stations in the Yangtze River valley region. When the index is positive, it is indicative of a typical flood year in the Yangtze River valley; when the index is negative, it is a drought year.

## 3. Results and discussions

3.1. EASM and Meiyu duration and their relation to aerosol pollution in the middle and lower reaches of the Yangtze River (M-LYR)

In Fig. 2a, EASM rainfall index of the 1961–2009 calculated by the formula (1) is presented. The positive correlation between EASM index and the duration of Yangtze River Meiyu (plum rain) is found with ~0.55 of the explained variance, suggesting that the duration of the Meiyu period is an important indicator of EASM strength. The relationship between them is widely used to reflect the drought/flood status of the Yangtze River valley (Liu et al., 2006; Zhou et al., 2009a). Under the weak monsoon years, the longer retention of MeiYu and floods in the drainage area of Yangtze River is found. On the contrary in the strong monsoon year, Meiyu duration is short, and drought is found in the Yangtze River basin.

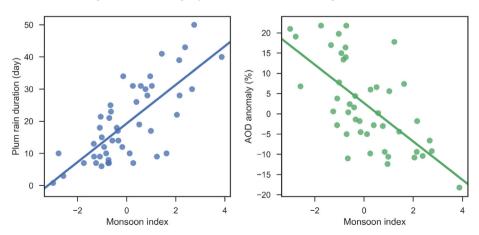


Fig. 2. Correlation between the summer monsoon index and plum rain duration (number of rainy days) in the Yangtze River Valley (a); and AOD anomaly (b).

The correlation between EASM index and the aerosol pollution (AOD anomaly relative to mean of 1961–1989) is illustrated in Fig. 2b. Negative correlation between EASM index and AOD departures is also found significantly with ~0.41 of the explained variance, suggesting that weak monsoon year generally corresponds to strong aerosol pollution in the M-LYR region. Conversely, strong EASM years normally less pollution condition.

The intensity of cross-equatorial monsoon significantly affects the amount and distribution of monsoon rainfall, and the cross-equatorial air flows are important components of tropical atmospheric circulations. It plays an important role in the exchange of water vapor, mass, momentum between southern and Northern Hemisphere. It will compensate for the aerosol induced energy imbalance between the Northern and Southern Hemispheres (Bollasina et al., 2011). Many studies have shown that a significant positive correlation is found between intensity of cross-equatorial flow and summer monsoon intensity. The stronger of cross-equatorial flow, then the stronger the summer monsoon will be and vice versa (Li et al., 2004). Research has shown that the cross-equatorial flow near 115°E is a major point of airflow for the East Asian monsoon, and is also the main source of flow impacting the Mei-yu, i.e. monsoon precipitation in East Asia (Wang and Li, 1982). According to Eq. (1), the monsoon drought/flood index (I) is calculated. By choosing the typical flood years of the Yangtze River valley (1973, 1980, 1983, 1996, 1998, 1999; I > 2) and less-rain drought years (1963, 1966, 1972, 1978, 1988; I < -1), synthetic analysis is separately conducted, producing an analysis of the meridional circulation profile of crossequatorial airflow near 115°E with the integrated data of flood years and drought years (Fig. 3a and b). It is seen from 3a, there is a strong monsoon anti-Hadley cell along the 115°E, whose main closed ring is located under 700 hPa. The monsoon airflow crosses the equator at a low altitude, moving forward to around 50°N. Subsequently, Plum rains residence time is short, AOD is a high value for the year, and the Yangtze River valley is in a less-rain drought year. In contrast, in weak monsoon years, the monsoon anti-Hadley cell at 115°E is notably weak. Its major feature is that the anti-Hadley cell splits into two closed rings in the vertical direction. Due to the split of the circulation ring, the low-level circulation becomes weak, the cross-equatorial airflow remains more south than in less-rain drought years, and maintains near 30°N. Then, strong updraft is formed near 30°N, causing the rain belt to persist over the Yangtze River valley (Figs. 3b), conducive to the rain band stranded in the Yangtze basin, Yangtze River in Meiyu period of delay, and may not conducive to aerosol stranded in middle and lower reaches of the Yangtze River, the AOD low-value years, which is beneficial to the development of strong rain (Fig. 3b). The inter-decadal variability of summer rainfall and temperature over East Asia were found to be largely influenced by the change of sea surface temperature and enhancement of a Hadley cell (Wang, 2001; Zhou et al., 2009b).

## 3.2. The changes in weather condition is also closely linked with change in EASM

The most direct affecting factor to aerosol pollution is unfavorable weather conditions during the adjacent year or certain period of time without much change in emissions. PLAM is one such index that establishes a link between aerosol pollution and unfavorable weather conditions (Wang et al., 2012; Wang et al., 2013; Zhang et al., 2009b). The higher the value of PLAM, the stronger the unfavorable weather condition and the stronger the contribution to the formation of local and regional aerosol pollution. Time series comparative analysis between the anomaly changes of PLAM and the EASM index exhibits a significant negative correlation since 1960's with ~0.70 of the explained variance (Fig. 4). Previous studies have shown that the EASM had a clear connection with the "southern China flood and northern China drought" rainfall pattern (Hu, 1997a; Wang, 2001; Yu et al., 2004; Zhou et al., 2009b). The pollution-related weather conditions in the area of our study were also affected by the rainfall duration and distribution.

During historic flood years of 1968, 1970, 1973, 1975, 1980, 1982, 1983, 1987, 1989 1993, 1995, 1996, 1999 and 2002, even a more strong negative correlation between PLAM and EASM index is found with ~0.80 of the explained variance. Conversely, during the strong EASM

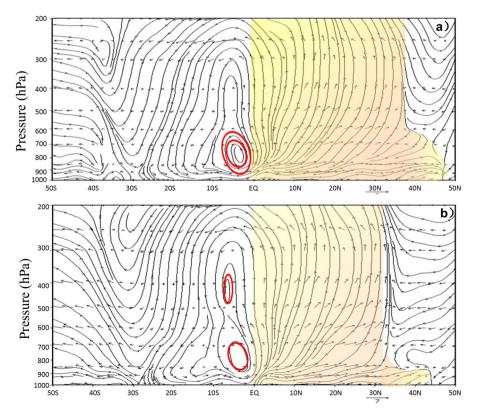


Fig. 3. Meridional circulation profile analysis at 115°E for less rainfall years (a); and water-logging years (b).

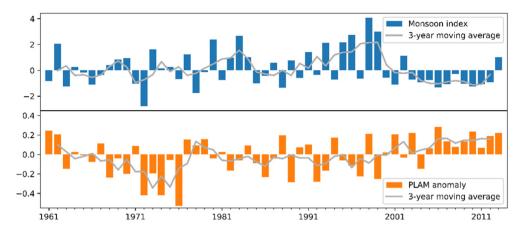


Fig. 4. Time series of summer (July) PLAM index anomaly in the M-LYR and the summer monsoon index from 1961 to 2014.

and drought years of 1961, 1967, 1971, 1978, 1988, 1990, 1994, 2000, 2001, 2003, 2005, 2006, 2007 and 2009, a stronger negative correlation between PLAM and EASM index is found with ~0.80 of the explained variance. This may result in the Yangtze River Delta region polluted weather conditions from the beginning of the late 1970's continue to slip, and the resulting aerosols increased. Simulation experiment shows that compared the difference of atmospheric conditions in East Asia from Europe, adequate water vapor in Summer could help to enhance the aerosols with moisture absorption (including sulphate) and hygroscopic growth in the troposphere, which will enhance its optical depth and corresponds to the direct radiative forcing (Li et al., 2012; Li et al., 2014). Thus it can be seen, despite the East Asian monsoon variability on China's regional distribution of aerosol concentrations and a significant and multiple mechanisms of influence, still the two factors are very important that the distribution of aerosol concentrations

depends on the emission sources and atmospheric conditions (Yu et al., 2012). To distinguish the relative contribution between emissions and atmospheric conditions on aerosol pollution changes is related to various government departments relating to the pollution control, and issues of concern to the scholars and the public.

3.3. The relative contribution of meteorological conditions to the interdecadal changes of aerosol pollution in M-LYR under climate change

The time series of PLAM and AOD in the M-LYR is presented in Fig. 5. Remarkable inter-annual variability of PLAM and AOD are found since 1960's. In order to get the inter-decadal change of them, a cubic splines function (CSF) polynomial filter is applied to remove off the inter-annual disturbance (Zhang et al., 1995). The generally synchronizing interdecadal changes of AOD and PLAM behave a relatively large decrease

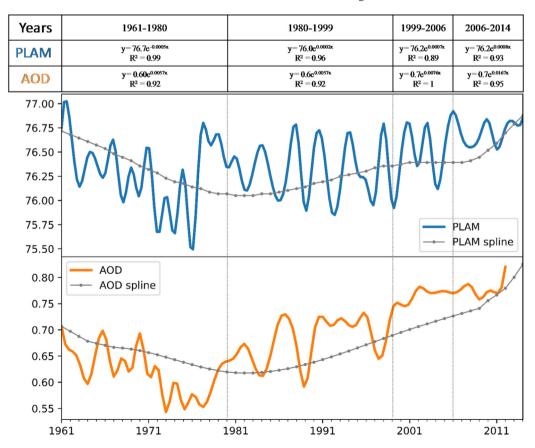


Fig. 5. Time series of summer PLAM index and its climate trend by PLAM\_spline, and AOD variability and its climate trend by AOD\_spline in M-LYR of China from 1961 to 2014.

**Table 1** 1961–2014 inter-decadal variations of PLAM (pollution meteorological condition index).

Period	years	Starting future $\alpha$	Change trend β	Continuous cycle λ (year)	PLAM contribution $\delta_{\text{C-PLAM}}$
I	1961-1980	76.74	$-5.0 \times 10^{-4}$	19	-73.3%
II	1980-1999	75.99	$2.0 \times 10^{-4}$	19	28.9%
III	1999-2006	76.24	$2.0 \times 10^{-4}$	7	10.7%
IV	2006-2014	76.25	$8.0 \times 10^{-4}$	8	48.9%

from 1961 to 1980, a large rise between 1980 and 1999, a period of slow rise or maintenance from 1999 to 2006, and a relatively rapid rise between 2006 and 2014. This gradual worsening of the weather conditions and aerosol pollution during the last three of these four periods in M-LYR normally coincides with the gradual weakening of the EASM since the 1970s (Hu, 1997b; Yu and Zhou, 2007; Yu et al., 2008; Zhou et al., 2009a; Zhou et al., 2009b). On the basis of the CSF, an e-exponential fitting of PLAM variation in each period since 1960's is presented as follow:

$$Y_{PLAM} = \alpha e^{\beta x} \tag{2}$$

The explained variance  $(r^2)$  among the 4 periods are 0.99, 0.96, 0.88 and 0.93, respectively, significant level exceeds 0.001. Among these parameters,  $\alpha$  represents the starting future of data,  $\beta$  denotes the change trend in data period,  $\lambda$  represents of continuous cycle of data (Wei, 1999). The amplitude of vibration in each stage of the data gives by formula (3):

$$\delta c = \alpha (e^{\beta \lambda} - 1) \tag{3}$$

All the PLAM data during the 4 periods are listed in Table 1.

Still on the basis of formula (3), inter-decadal variations of AOD are also obtained from 1961 to 2014 (Table 2). The explained variance  $(r^2)$  among the 4 periods are also high with 0.98, 0.92, 0.99 and 0.95, respectively (also marked in Fig. 5), with the significance level exceeding 0.001. The relative contribution from meteorological condition (MC) and anthropogenic emissions (AE) to each period of the four are presented in Table 3.

From Tables 1 to 3, one can see:

1) During the period of 20 years for 1961–1980 (period I, shown in Fig. 5), decreasing of the PLAM index is highly synchronized with changes in AOD. Since this stage, one can assume the contribution of anthropogenic emission on the aerosol pollution is very small, and all the aerosol pollution changes can be considered as a result of changes in weather conditions, because China has not yet opened up to the world during this period, the level of various anthropogenic emission activities is extremely low. The ratio (called RC, 10.8) of variations in AOD ( $\delta$ -C-AOD, -7.9%) and weather condition ( $\delta$  C-PLAM, -73.3%) can be considered as an inherent coefficient between the changes in meteorological conditions and aerosol pollution in this region. So, the contribution of anthropogenic emissions on aerosol pollution for other three periods can be approximately estimated as follow:

$$\delta_{\text{AOD-emissions}} = \delta_{\text{AOD-obs}} - (\delta_{\text{C-PLAM}} \times \text{RC}) \tag{4}$$

**Table 3**Inter-decadal variations of influence of meteorological condition (MC) and anthropogenic emissions (AE) contributing to the pollutions for 1961–2014.

Period	Years	PLAM $\delta_{\text{C}}$ -PLAM	AOD $\delta_{\text{C}}$ -AOD	RC	AE contribution (%)	MC contribution (%)
I	1961-1980	-73.3%	-7.9%			(100%)
II	1980-1999	28.9%	6.1%	3	49%	51%
III	1999-2006	10.7%	5.3%	4	75%	25%
IV	2006-2014	48.9%	8.7%	3.5	40%	60%

- 2) During the period of 1980–1999 (period II), The increase of AOD is around 6.1% (Table 3), Of the 6.1%, 3.0% can be considered to come from anthropogenic emission, showing that ~49% of aerosol pollution increases indicated by AOD during the summertime can be attributable to anthropogenic emission in the M-LYR area during the 20 years after the 1980. There was about 51% of the change in aerosol pollution coming from worsened weather conditions under climate change.
- 3) During the period of 1997–2007 (Period III), the relative contributions from anthropogenic emission and meteorological condition to aerosol pollution were 75% and 25%, respectively.
- During the period of 2007–2014 (Period IV), the rise of PLAM is very similar to the AOD rise. The estimated meteorological contribution on the aerosol pollution is around 60%.

#### 4. Conclusion

The interdecadal changes in meteorological conditions closely related to summer aerosol pollution in M-LYR were highly impacted by the change of EASM, and have worsened gradually since the 1980s, accompanied by similar changes in aerosol pollution in this area.

During the weak monsoon years with lower PLAMs (better weather conditions), the monsoon related anti-Hadley cell at 115°E is notably weak and be maintained near 30°N; the weather conditions are conducive to the maintenance of the rainband in M-LYR, and this area enters in Meiyu period of delay, accompanied by the emergence of the flood and lower aerosol pollution. Conversely, during the stronger monsoon years with higher PLAMs (worsened weather conditions), the monsoon related anti-Hadley cell at 115°E is also stronger and the airflow that crosses the equator at a low altitude move forward to around 50°N; Subsequently, Plum rains residence time is short in the M-LYR, accompanied by the emergence of the drought and higher aerosol pollution.

In recent 10 years since 2006, the meteorological condition in summertime in the M-LYR is the worst 10 years since 1960, accompanied by around 49% increase of the summer aerosol pollution. ~60% of this increase pollution can be attributable to worsening meteorological conditions, and the rest of 40% comes from the increase of anthropogenic emissions during recent years.

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**Table 2** 1961–2014 AOD inter-decadal variations.

Period	Years	Starting future $\alpha$	Change trend $\beta$	Continuous cycle λ(year)	AOD δ <sub>C</sub> -AOD	AOD anomaly peak value
I	1961-1980	0.70	$-6.3 \times 10^{-3}$	19	-7.9%	
II	1980-1999	0.60	$5.7 \times 10^{-3}$	17	6.1%	> 0.10
III	1999-2006	0.67	$7.6 \times 10^{-3}$	10	5.3%	> 0.10
IV	2006-2014	0.70	$16.7 \times 10^{-3}$	7	8.7%	>0.14

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